# Characterization and Classification of Unprocessed Solid Radioactive Wastes Stored at the Central Radioactive Waste Processing and Storage Facility of Bangladesh Atomic Energy Commission

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## Abstract

The presence of probable artificial radionuclides and their activity concentration levels in unprocessed solid radioactive waste stored at the interim storage area of Central Radioactive Waste Processing and Storage Facility (CWPSF), Institute of Nuclear Science and Technology (INST), Atomic Energy Research Establishment (AERE), Savar, Dhaka, Bangladesh were investigated. The study was conducted for characterization and quantitative classification of the unprocessed radioactive wastes to decide the subsequent management steps of the wastes. A total of twenty one samples were collected from three different storage drums. The samples were analyzed by gamma spectrometry technique using a high purity Germanium (HPGe) detector with 20% relative efficiency. Besides, airborne particles inside the CWPSF were collected using a staplex high volume air sampler with glass fiber filter. The filters were also analyzed by gamma spectrometry technique. Gamma spectral analysis of the collected samples indicated the presence of two artificial radionuclides; $^{60}$Co and $^{137}$Cs. The activity concentrations of $^{60}$Co were found in the range 33.15 ± 8.74 Bqkg$^{-1}$to 702.61 ± 48.36 Bqkg$^{-1}$with an average of 175.17 ± 22.82 Bqkg$^{-1}$. The activity concentration of $^{137}$Cs was found 39.37 ± 9.14 Bqkg$^{-1}$in only one sample. No artificial radionuclide was detected in any of the filters used for air sampling in the facility except the naturally occurring radionuclides. Radioactive waste characterization and classification were conducted according to IAEA Safety Standard GSG-1. The results of the current study would help to decide the final end-point of these wastes.

***Keywords**:* Radioactive waste, HPGe detector, Activity concentration, Air sampler, Characterization and classification of radioactive waste

## 1. Introduction

Once created, radioactive waste may undergo several processing steps such as pre-treatment, treatment, conditioning, storage and disposal depending on the type of waste and strategy for its management [1]. The guidance for treatment and conditioning of radioactive waste is based on data obtained on waste material characterization. Characterization of radioactive waste gives important waste material parameters and enables its classification according to national as well as international regulations. Although classification schemes are country-specific, there is a generic consensus that conditioning methods (e.g. immobilization and packaging) and end points (e.g. storage and disposal) depend on the level of radioactivity and radionuclide lifetime. Radioactive waste processing routes are specified herein using the new International Atomic Energy Agency (IAEA) radioactive waste classification scheme which is based on long-term safety of wastes [2]. On the other hand, classification of the wastes is also very helpful throughout their management phases from generation through collection, segregation, treatment, conditioning, storage, transportation to final disposal. Generally, wastes are remaining three states: solid, liquid and gaseous. The IAEA classified different types of radioactive waste as Exempt waste (EW), Very short lived waste (VSW), Very low level waste (VLLW), Intermediate level waste (ILW) and High level waste (HLW) [1].

The importance of the safe management of radioactive waste for the protection of human health and the environment has been long recognized. When the radiological hazards associated with radioactive waste is negligible, then the waste can be released from nuclear regulatory control in accordance with established clearance levels in a country. Clearance levels are a set of values, established by the regulatory body in a country or state, expressed in terms of activity concentrations and/or total activities, at or below which sources of radiation can be released from nuclear regulatory control [3].

In Bangladesh solid and liquid radioactive wastes are being generated from the operation and maintenance of BAEC TRIGA Research Reactor (BTRR) and from usage of radioisotopes in research and education, medicine, agriculture, industrial practices etc. The radioactive wastes arising in the country are generally spent ion-exchange resins, graphite from research reactor, contaminated vials, hand gloves, plastic syringes, shoe-covers, protective cloths, plastic and metallic wares, spent and disused sealed radiation sources (DSRS) etc. In order to process and safe storage of radioactive wastes on interim–basis until final disposal, a Central Radioactive Waste Processing and Storage Facility (CWPSF) has been established at INST, AERE, Savar, Dhaka. This facility is divided into several areas namely; waste accepting area, segregation area, conditioning area and interim storage area as well as facility has necessary equipment for processing the radioactive

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wastes. The radioactive wastes are stored at the facility in 200 L drums as shown in Fig. 1. Radioactive wastes collected mainly from research reactor, radioisotope production facility, nuclear medicine facilities and various research organizations are stored at the storage area of the CWPSF. Sometimes the waste packages received by the facility are unidentified regarding their physical and chemical form, radionuclide content, and activity level etc.

**Fig. 1:** Storage area of processed and unprocessed radioactive wastes at the CWPSF

The objective of the current study is to characterize(e.g., the measurement of physical parameters, radiation dose levels, the identification of radionuclides and the measurement of activity content, etc.) the unprocessed solid radioactive wastes collected mainly from research reactor to decide their final end-point in the subsequent management steps. In order to characterize and classify the solid radioactive waste attempt has been made to segregate the wastes depending on their physical states as well as identification and quantification of artificial radionuclides, activity concentration level etc. by gamma ray spectrometry system using a High Purity Germanium (HPGe) detector. In the present study, a total of 21 samples were analysed collected from 3 (three) different drums stored at the interim storage area of the CWPSF.

## 2. Materials and Method

### *2.1 Sample Collection and Processing*

In the present study, a total of 21 solid radioactive waste samples were collected from three different drums stored at the interim storage room of CWPSF (as given in Table 1). The waste drums were stored for about 9-23 years at the CWPSF for the decay of radionuclide contents in the wastes.

**Table 1:** Collected solid radioactive waste samples

| Waste-drum ID | Waste collection date at CWPSF | Sample collection date | Sample ID | Weight of the collected waste (gm) | Max. surface dose rate (μSv/h) | Surface contamination level (Bq/cm2) |
|---|---|---|---|---|---|---|
| Rs_Us_Un_21 | 18.04.1993 | 29.10.2015 | USRW-01 | 12.20 | 0.27 | 0.40 |
| | | 14.06.2016 | USRW-02 | 25.55 | 0.33 | 0.45 |
| | | | USRW-03 | 23.01 | 0.19 | 0.26 |
| | | | USRW-04 | 58.47 | 0.21 | 0.38 |
| | | | USRW-05 | 33.39 | 0.11 | 0.26 |
| | | | USRW-06 | 25.21 | 0.22 | 0.40 |
| | | | USRW-07 | 17.75 | 0.33 | 0.64 |
| | | | USRW-08 | 30.45 | 0.64 | 0.26 |
| | | | USRW-09 | 22.49 | 0.21 | 0.31 |
| Rs_Us_Un_20 | 18.04.1993 | 27.06.2016 | USRW-10 | 27.07 | 0.30 | 0.52 |
| | | | USRW-11 | 32.26 | 0.44 | 0.41 |
| | | | USRW-12 | 60.10 | 0.32 | 0.29 |
| | | | USRW-13 | 46.42 | 0.30 | 0.40 |
| | | | USRW-14 | 20.67 | 0.17 | 0.42 |
| | | | USRW-15 | 19.29 | 0.26 | 0.47 |
| | | | USRW-16 | 21.05 | 0.29 | 0.57 |
| | | | USRW-17 | 32.35 | 0.19 | 0.40 |
| Rs_Us_Un_14 | 16.01.2008 | 28.07.2016 | USRW-18 | 75.71 | 0.13 | 0.38 |
| | | | USRW-19 | 20.95 | 0.52 | 0.52 |
| | | | USRW-20 | 35.52 | 0.15 | 0.42 |
| | | | USRW-21 | 21.49 | 0.24 | 0.41 |

These wastes were generated from the operation and maintenance of BAEC TRIGA Research Reactor (BTRR). The wastes were mainly ion-exchange resins, hand gloves, plastic shoe-covers, protective cloths, plastic and metallic wares etc. During the collection of the samples the gamma radiation dose rate and contamination level on the outer surface of the drums were measured by different types of radiation measuring equipment. Then the drums were opened and the wastes were segregated carefully depending on their physical states. Afterwards, some samples (e.g., hand gloves, shoe-covers, protective cloths, etc.)were taken randomly from the segregated wastes and subsequently poured in to the plastic containers and marked with identification parameters such as waste package/drum number, sample ID, collection date, etc. The gamma radiation dose rate and contamination level on the surface of the each of the collected samples were measured. The personnel involved in this work took appropriate radiation protection measures (such as lead aprons, lead goggles, disposable hand gloves, musk, protective shoes, etc.) to protect themselves from radiation hazards. They also used personal dosimeters to measure the absorbed dose during the work. Table 1 shows the detail information on the sample collection and collected samples used for the present work.

A Staplex high volume Air Sampler (with glass fibre filter of 4″ diameter and with 1 μm pore size) was used to trap the airborne radioactive particles present in the air of the storage area. The air sampler was placed at the height of 1 m above from the ground and was run for about 15-30 minutes. The samples were collected after passing approximately 10-19 $m^3$ of air through the filter papers. A High Purity Germanium (HPGe) detector with 20% relative efficiency incorporated with relevant accessories was used to identify and quantify the gamma emitting radionuclides trapped in this filter. Fig. 2 shows the air sampler and filter used for the measurement of air quality in storage area.

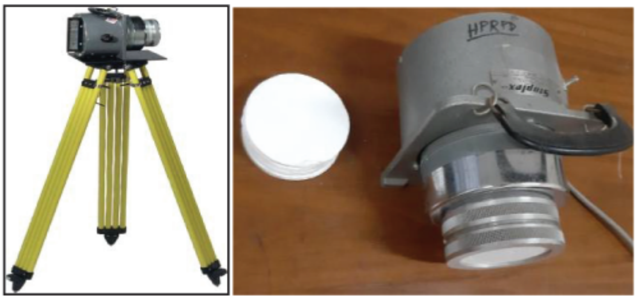

**Fig. 2:** High volume air sampler and filter used for trapping the suspended radioactive particles in the storage area

### 2.2 Sample Analysis

The detection and measurement of radionuclides in the samples were carried out by gamma spectrometry system using a vertical coaxial cylindrical high purity germanium (HPGe) detector of 172 $cm^3$ active volume and 20% relative efficiency. The p-type HPGe detector was supplied by CANBERRA (Model-GC2018). The detector was coupled to a 16 k-channel computer analyser. The analysis was carried out using Genie 2000 software, which matched various gamma energy peaks to a library of possible radionuclides. All the samples were counted for 10 ks. Before the measurement of the samples, the gamma background at laboratory site was determined with an identical empty plastic container used in the sample measurement. The gamma spectral analysis indicated that artificial radionuclides like $^{137}Cs$ and $^{60}Co$ were present in some of the analysed samples (as shown in Fig. 3). The gamma ray lines of energies 1173.24 keV (99.85%) and 1332.508 keV (99.99%) were used for the identification of $^{60}Co$, whereas, $^{137}Cs$ was detected by using gamma ray line of energy 661.659 keV (84.99%) [4].

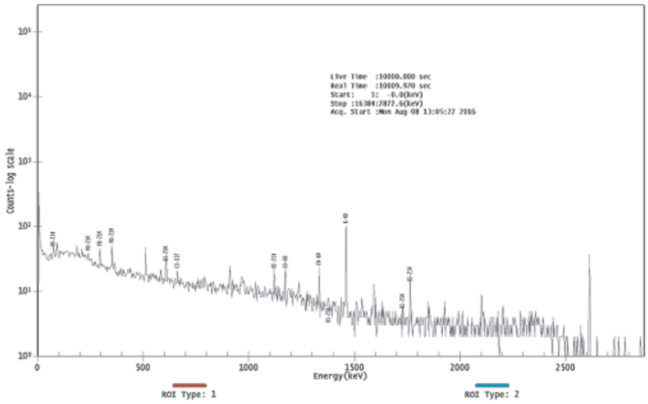


**Fig. 3:** A typical spectrum of the analysed sample

The error in the measurement was reported as combined standard uncertainties with coverage factor, k=2which corresponds to the 95% confidence level. The efficiency calibration of the detector was performed by homogeneously incorporating of standard solutions of $^{152}Eu$ into inactive matrices ($Al_2O_3$). The standard was prepared by pipetting a volume of the solution on to the matrices and subsequent drying in sample drying oven. In order to ensure that the radionuclides were homogeneously distributed in the matrices, the prepared standard was carefully mixed before measurement [5-6]. The energy calibration of the detector was performed by $^{137}Cs$ and $^{60}Co$ point sources.

### 2.3 Measurement of Activity Concentration

Activity concentrations of the artificial radionuclides in the collected waste samples were estimated by considering the net count rate, counting efficiency and emission rate of certain radionuclides and weight of sample as per the following equation [7]:

$$A = \frac{Net\ Count\ Rate}{E_{ff} \times I_{\gamma} \times w} \quad (1)$$

where, $A$ is activity concentration ($Bqkg^{-1}$); *Net Count Rate*= Counts per second of sample - Counts per second of background; $E_{ff}$ *is* the counting efficiency of the HPGe detector; $I_{\gamma}$ is the emission probability and w is the sample mass (kg). In this study, the minimum detectable activity (MDA) of the gamma-ray measurement system was calculated by the following equation [8]:

$$MDA\ (Bq\ kg^{-1}) = \frac{1.645\sqrt{B}}{E_{ff} \times I_{\gamma} \times w \times T} \quad (2)$$

where, $B$, is the background counts over the region of interest for each radionuclide and $T$, is the measurement time in seconds.The MDA for different radionuclides of interest are given in Table 2.

**Table 2:** Minimum detectable activity for different radionuclides

| Radionuclides | Energy (keV) | MDA ($Bqkg^{-1}$) |
|---|---|---|
| $^{133}Ba$ | 356.1 | 4.20 |
| $^{137}Cs$ | 661.79 | 4.85 |
| $^{54}Mn$ | 834.98 | 0.49 |
| $^{56}Zn$ | 1115.74 | 3.13 |
| $^{152}Eu$ | 121.7817 | 4.61 |
| $^{60}Co$ | 1173.48 | 5.84 |
| $^{60}Co$ | 1332.71 | 5.22 |

## 3. Results and Discussion

As mentioned earlier in Table 1 that a total of 21 samples from 3 different drums (Rs_Us_Un_21, Rs_Us_Un_20 and Rs_Us_Un_14) were collected from the interim storage room of CWPSF and were analyzed by gamma spectrometry system. The analysis revealed that the radionuclides like, $^{60}Co$ and $^{137}Cs$ were present in some of the collected samples. The activity concentrations of the radionuclides in the wastes on waste collection date were estimated based on the present activity concentrations and half-life of the radionuclides in the wastes. The activity concentrations of the identified radionuclides along with their clearance levels for landfill disposal are shown in Table 3 while the Fig. 4 illustrates their graphical representation [8].

In Drum No. 21, the activity concentrations of $^{60}Co$ in the samples with ID USRW-01, USRW-02, USRW-03, USRW-06, USRW-07 and USRW-08 were found to be 198.22 ± 35 Bq kg$^{-1}$, 78.61 ± 15.21 Bq kg$^{-1}$, 702.61 ± 48.36 Bq kg$^{-1}$, 162.08 ± 22.21 Bq kg$^{-1}$, 162.33 ± 26.44 Bq kg$^{-1}$ and 33.15 ± 8.74 Bq kg$^{-1}$, respectively. On the other hand, no radionuclides were identified in samples USRW-04, 05& 09 and hence,mentioned as ND and the concentrations are stated as less than the minimum detectable activity (<MDA) in Table 3.

**Table 3:** Data for activity concentrations of artificial radionuclides along with their clearance levels in the solid radioactive waste samples collected from CWPSF

| Waste/Drum ID | Sample ID | Name of the identified radionuclides | Activity concentrations (Bq kg$^{-1}$) | | Clearance levels for landfill disposal (Bq kg$^{-1}$) [8] |
|---|---|---|---|---|---|
| | | | Present activity | At waste collection date in CWPSF | |
| Rs_Us_Un_21 | USRW-01 | $^{60}Co$ | 198.22 ± 35 | 3847 | 500 |
| | USRW-02 | $^{60}Co$ | 78.61 ± 15.21 | 1656 | |
| | USRW-03 | $^{60}Co$ | 702.61 ± 48.36 | 14821 | |
| | USRW-04 | ND* | <MDA** | – | – |
| | USRW-05 | ND | < MDA | – | – |
| | USRW-06 | $^{60}Co$ | 162.08 ± 22.21 | 3418 | 500 |
| | USRW-07 | $^{60}Co$ | 162.33 ± 26.44 | 3421 | |
| | USRW-08 | $^{60}Co$ | 33.15 ± 8.74 | 702 | |
| | USRW-09 | ND | < MDA | – | – |
| Rs_Us_Un_20 | USRW-10 | ND | < MDA | – | – |
| | USRW-11 | ND | < MDA | – | – |
| | USRW-12 | ND | < MDA | – | – |
| | USRW-13 | ND | < MDA | – | – |
| | USRW-14 | ND | < MDA | – | – |
| | USRW-15 | ND | < MDA | – | – |
| | USRW-16 | ND | < MDA | – | – |
| | USRW-17 | ND | < MDA | – | – |
| Rs_Us_Un_14 | USRW-18 | $^{60}Co$ | 37.95 ± 6.2 | 117 | 500 |
| | USRW-19 | $^{137}Cs$ | 39.37 ± 9.14 | 48 | 1000 |
| | USRW-20 | $^{60}Co$ | 111.96 ± 20.19 | 346 | 500 |
| | USRW-21 | $^{60}Co$ | 89.64 ± 13.90 | 277 | 500 |

ND* = Not detected, MDA** = Minimum detectable activity

On the other hand, no artificial radionuclides were detected in any of the samples collected from Drum No. 20, hence are reported as ND in Table 3. Consequently, the activity concentrations are denoted as <MDA of the counting

system. As the activity concentrations are less than the MDA of the counting system, therefore, the waste in Drum No. 20 does not require any further processing and can be disposed to the environment upon approval from the regulatory authority "Bangladesh Atomic Energy Regulatory Authority (BAERA)".

Moreover, in Drum No. 14, the activity concentrations of $^{60}$Co in the samples USRW-18, USRW-20 & USRW-21 were found to be 37.95 ± 6.2 Bq kg$^{-1}$, 111.96 ± 20.19 Bq kg$^{-1}$and 89.64 ± 13.90 Bq kg$^{-1}$, respectively. Besides, the activity concentration of $^{137}$Cs in the sample USRW-19 was observed as 39.37 ± 9.14 Bq kg$^{-1}$.Based on the activity concentration, the classification scheme stated in General Safety Guide: GSG-1indicating that the waste in Drum Rs_Us_Un_21 and Rs_Us_Un_14 are belongs to low level radioactive waste (LLW) [2].

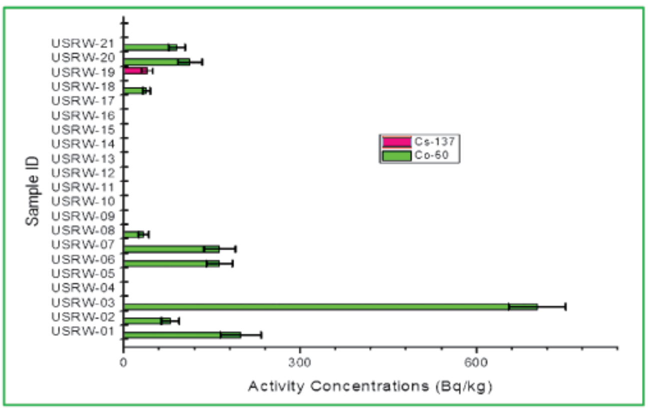


**Fig. 4(a):** Graphical representation of activity concentrations of artificial radionuclides present in the analyzed samples

Besides, amongst the analyzed samples, the sample with waste ID USRW-03 in the waste drum Rs_Us_Un_21 has the present activity concentration of 702.61 ± 48.36 Bq kg$^{-1}$(for $^{60}$Co), which is slightly higher than the clearance level (500Bq kg$^{-1}$) for landfill disposal [3]. On the other hand, the estimated concentrations (at the waste collection date in CWPSF) for almost all the samples (except USRW-04 and USRW-05) of drum no. Rs_Us_Un_21 were clearly higher than that of the clearance levels for landfill disposal. Due to delay and decay of approximately 23 years at CWPSF, the activity concentrations of $^{60}$Coreach nearly below the clearance level for all the samples except USRW-03. Moreover, in order to guesstimate after how many years, the waste in drum Rs_Us_Un_21 can be cleared from regulatory control, a decay curve has been plotted and is shown in Fig. 4. It is observed that for decay of the maximum estimated activity concentration (for $^{60}$Co) of 14821 Bq kg$^{-1}$below clearance level may take approximately 26 years. Hence, the waste in drum Rs_Us_Un_21 has to be stored for about 3 years more to be cleared from regulatory control. As the activity concentration of the radionuclides in Rs_Us_Un_14 are far below their respective clearance levels for landfill disposal, hence, may be disposed to the environment upon the approval from regulatory body.

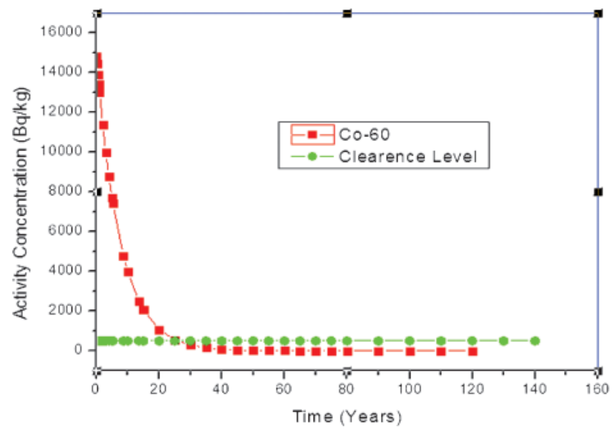


**Fig. 4(b):** Decay of $^{60}$Co activity concentration in the sample USRW-03

### *3.1 Airborne radioactivity monitoring*

Airborne particles inside the CWPSF were collected using a Staplex High Volume Air Sampler with glass fiber filter and analyzed by gamma spectrometry technique. The gamma spectral analysis of the filter samples indicated that no artificial radionuclides was detected in any of the collected samples except the naturally occurring ones(as shown in Fig. 5). Therefore, the results of the present study may provide useful information for ensuring safety of the radiation workers in the facility involved in radioactive waste management activities.

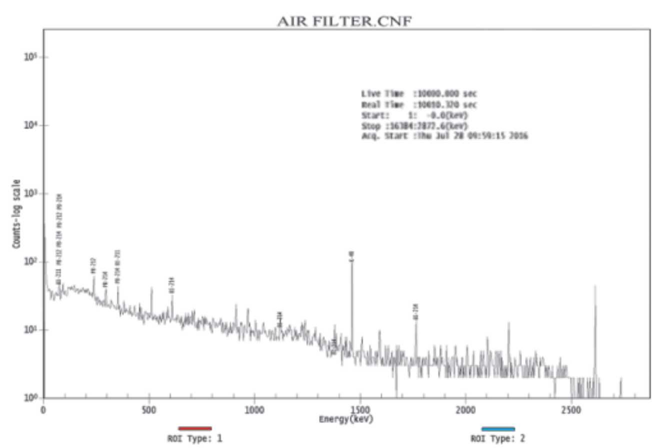


**Fig. 5:** A typical spectrum of the analyzed air sample

## 4. Conclusion

Identification of radionuclides and measurement of their activity concentration in 21 samples of 3 waste-drums stored at the CWPSF have been performed in the present study. The experimental result showed that almost all the samples collected from drums with Waste-Drum ID 14 and 21were contaminated with artificial radionuclides (except samples with ID: USRW-04,USRW-05, USRW-09 and USRW-21). Gamma spectral analysis of thecollected samples indicated the presence of two artificial radionuclides$^{60}$Coand $^{137}$Cs.As per the General Safety Guide: GSG-1, the activity concentration and classification scheme showed that the wastes under investigation belong to low level radioactive waste (LLW).Moreover, according to IAEA-TECDOC-855, the waste in drum no. Rs_Us_Un_21 have the activity concentration which is slightly higher than the clearance level for landfill disposal and has to be stored for more time to be cleared from regulatory control. Waste stored in the waste drum with IDRs_Us_Un_20canbe disposed to the environment upon approval from the regulatory authority as no artificial radionuclide was detected in any of the samples. The analysis of the filter samples used for identification and quantification the airborne radio activity at the workplace showed no artificial radionuclide in any of the filter samples except the naturally occurring ones.

The current research is confined at a limited number of samples of unprocessed solid radioactive wastes collected from three different containers stored at interim storage room of CWPSF in order to characterize the radioactive wastes and investigate the presence of artificial radionuclides along with their activity concentration. However, picture maybe different in different types of wastes depending on their physical states, radiological characteristics and quantities, radiation dose rates etc. Therefore, an extensive laboratory investigation is need to be carried out in order to get a clear picture of radioactivity contamination in these wastes for their classification as well as for deciding next management steps.

## References

[1] International Atomic Energy Agency (IAEA), Predisposal Management of Radioactive Waste from Nuclear Power Plants and Research Reactors, IAEA Safety Standards Series No. **SSG-40**, 38-47 (2016).

[2] International Atomic Energy Agency (IAEA), classification of radioactive waste, General Safety Guide, No. **GSG-1**, 11-14, 30-32, 41 (2009).

[3] International Atomic Energy Agency (IAEA), Clearance levels for radionuclides in solid materials, IAEA-TECDOC-**855**, 21-22 (1996).

[4] http://www.lnhb.fr/nuclear-data/nuclear-data-table.

[5] A. Samad, A. Haydar, I. Ali, D. Paul, M.R. Bhuiyan and SM. Islam, A study on the radioactivity level in raw materials, fnal products and wastes of the phosphate fertilizer industries in Bangladesh, J Environ Prot., **3 (10)**, 1393-1402 (2012).

[6] S. Harb, K.S. Din and A. Abbady, Study of Efficiency Calibrations of HPGe Detectors for Radioactivity Measurement of Environmental Samples, Proceedings of the 3rd Environmental Physics Conference, Aswan, 207-218 (19-23 February 2008).

[7] S.S. Islam, M.I. Ali, M.A. Haydar, M.M. Hasan, B. M.R. Faisal, S. Karmaker, M.A. Shariff, M. Ali, D. Paul and S M. A. Islam, Background gamma radiation mapping in forest ecosystem of Bangladesh: A study on the radioactivity distribution in the national reserve forest of Gazipur, Radiat Prot Environ, **40,** 73-83 (2017

[8] K. Asaduzzaman, F. Mannan, M.U. Khandaker, M.S. Farook, A. Elkezza, Y.M. Amin, S.Sharma and H.A. Kassim, Assessment of Natural Radioactivity Levels and Potential Radiological Risks of Common Building Materials Used in Bangladeshi Dwellings, PLOS ONE, **10(10)**, 1-16 (2015).